Mixing in porous media

P. de Anna, post-doc fellow at MIT
(Civil and Environmental Engineering Department)
mixing
mixing

two or more given substances, originally segregated into different volumes of space, tend to occupy the same volume due to mixing processes (e.g. Ottino, 1989)

a mixture is a transient state between the initial segregation of the constituents and their ultimate homogeneity (Villermaux and Duplat., 2003)
mixing is done ONLY by molecular diffusion

The individual random walk (Brownian motion) of molecules makes an initial compact cloud to increase its volume.

The mass flux can be measured or theoretically computed: it follows the Fick's law:

\[ \frac{\partial c}{\partial t} = \nabla \cdot J = D \nabla^2 c \]

The cloud size grows as \( \sqrt{\sigma^2} = \sum_i^N \sqrt{(x_i(t) - \mu)^2} = \sqrt{Dt} \)
mixing with advection

Heterogeneous flows enhance the solutes interface...

...where diffusion takes place

mixing: immiscibility

Not all fluids are miscible!
porous media
are ubiquitous in nature and industry
porous media

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microtomography - Oregon State University, Wildenschild Research Group
porous media

are ubiquitous in nature and industry
Porous medium

1. “[...] a portion of space occupied by heterogeneous or multiphase matter. [...] The solid phase is called solid matrix”

2. ”[...] the specific surface of the solid matrix is relatively high [...] the various openings (the pores) comprising the void space are relatively narrow“

3. ”[...] at least some of the pores comprising the void space should be interconnected. “

J. Bear Dynamics of fluids in Porous Media, 1972
Transport at the Darcy scale description
dye within a sand box

continuum (macroscopic) vs. discrete (pore scale) approach
Microorganisms and chemicals influence environmental dynamics. Large-scale consequences result from the interactions occurring at microscale...
Conservation laws
Momentum conservation: the flow equation

Navier-Stokes equations for a Newtonian fluid:

\[ \rho \left( \frac{\partial \mathbf{u}}{\partial t} + \mathbf{u} \cdot \nabla \mathbf{u} \right) = -\nabla p + \mu \nabla^2 \mathbf{u} + \mathbf{f}. \]

where

- \( \rho \) is the fluid density
- \( \mathbf{u} \) is the fluid velocity
- \( p \) is the local pressure
- \( \mu \) the dynamics viscosity of the fluid
Momentum conservation: the flow equation

Navier-Stokes equations for a Newtonian fluid:

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To characterize the flow we define the Reynolds number

\[
\text{Re} = \frac{\text{inertial forces}}{\text{viscous forces}} = \frac{\rho \mathbf{u} L}{\mu}
\]

If \( \text{Re} \ll 1 \), in stationary conditions the N.S. equation reduces to the **Stokes** equation:

\[
\nabla p = \mu \nabla^2 \mathbf{u}
\]
Momentum conservation: the flow

Chaotic incompressible flow $Re > 10^2$

Momentum conservation: the flow

Chaotic incompressible flow $Re > 10^2$


Pore scale flow $Re \sim 10^{-3}$

Momentum conservation: flow intermittency - 2D

Momentum conservation: flow intermittency - 2D

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Momentum conservation: flow intermittency - 2D

\[ \Delta v_p = v_p(t + \tau) - v_p(t) \]


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Momentum conservation: flow intermittency - 3D

\[ \chi_a = \left\langle \frac{(a_p(t) - \langle a(t) \rangle)(a_p(t+\tau) - \langle a(t+\tau) \rangle)}{\sigma_a(t)\sigma_a(t+\tau)} \right\rangle \]
Momentum conservation: flow intermittency - 3D

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Momentum conservation: flow intermittency


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Flow through porous media: the Darcy scale

Stokes flow

\[ \nabla p = \mu \nabla^2 \mathbf{u}, \]

\( p \) is sol. of heat equation

Darcy's law

\[ Q = -\frac{k A (P_b - P_a)}{\mu L}, \]

- \( Q \) is the total flow
- \( k \) is the medium permeability
- \( P_b \) and \( P_a \) are the inlet and outlet pressures
- \( L \) the medium length
- \( \mu \) the fluid viscosity
Flow through porous media: the Darcy scale

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- \( \mu \) the fluid viscosity

Generalization to fields:

\[ \mathbf{q} = \frac{k}{\mu} \nabla p \]

\( p \), is sol. of heat equation
Transport of a scalar quantity $c$

Mass conservation at each spatial location:

\[
\frac{\partial c}{\partial t} = \nabla \cdot (-uc + \vec{J}) = \nabla \cdot (-uc + D\nabla^2 c)
\]

the flux is due to advection and diffusion and $\vec{J} = D\nabla c$, $D$ is the Diffusion coefficient (Fick’s law).

The Péclet number $Pe = \frac{u\lambda}{D}$ quantifies the characteristic advection to diffusion time.
Transport of a scalar quantity $c$ - the pore scale

velocity field (Stokes flow)  

injected concentration field

P. de Anna et al. *Phys. Rev Lett.* 2013
Transport of a scalar quantity $c$ - the pore scale

velocity field (Stokes flow)

injected concentration field

spreading of the front vs. mixing at the front

P. de Anna et al. *Phys. Rev Lett.* 2013

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Transport of a scalar quantity \( c \) - the Darcy scale

Darcy flow field \( q \) for a given \( k \)

\[
q = \frac{k}{\mu} \nabla p
\]
Transport of a scalar quantity $c$ - the Darcy scale

Darcy flow field $q$ for a given $k$

$$q = \frac{k}{\mu} \nabla p$$

plume concentration field for different $k$

Transport of a scalar quantity $c$
Transport of a scalar quantity $c$
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(juanesgroup.mit.edu/pdeanna) pietrodeanna@gmail.com

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Transport of a scalar quantity $c$

Lamellar description: the concentration distribution across the front is Gaussian

$$c(\zeta, t) = \frac{c_0 \exp \left[ -\frac{\zeta^2/s^2(t)}{1+4\tau(t)} \right]}{\sqrt{1 + 4\tau}}$$

$$\tau = \int_0^t dt' \frac{D\rho(t')}{s_0^2}, \quad \rho = \frac{L(t)}{L_0}, \quad \theta = \frac{c_0}{1 + 4\tau}$$

Existence of two regimes: stretching and coalescence of lamellae.
Transport of a scalar quantity $c$

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$$p(c|\theta) = p(x) \left( \frac{dc}{dx} \right)^{-1} = \frac{1}{c \sqrt{\ln(\theta/\epsilon)} \ln(\theta/c)}$$
Transport of a scalar quantity $c$

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$$p(c|\theta) = p(x) \left( \frac{dc}{dx} \right)^{-1} = \frac{1}{c \sqrt{\ln(\theta/\epsilon) \ln(\theta/c)}}$$

$$\rightarrow \quad p(c, t) = \int_{c,0}^{c_0} d\theta \frac{p_m(\theta, t)}{c \sqrt{\ln(\theta/\epsilon) \ln(\theta/c)}}$$

Existence of two regimes: **stretching** and **coalescence** of lamellae.
The Advection-Diffusion-Reaction equation

Mass conservation:

\[ \frac{\partial c}{\partial t} = -\mathbf{v} \cdot \nabla c + D \nabla^2 c + r_c(\mathbf{x}, t) \]

\( r_c(\mathbf{x}, t) \) is the rate of consumption of \( c \) by the chemistry.

A.D.R. equations
Reactions

A chemical reaction is a process that leads to the transformation of one set of chemical substances to another.
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typical chemical equation: $\alpha A + \beta B \rightarrow \text{product}$
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A chemical reaction is a process that leads to the transformation of one set of chemical substances to another.

typical chemical equation: \( \alpha A + \beta B \rightarrow \text{product} \)

The speed at which a reaction takes place is studied by reaction kinetics. The rate depends on:

- concentrations
- Surface area available for contact between the reactants
- Pressure
- Activation energy
- Temperature
- presence or absence of a catalyst
Reactions

How to quantify the rate of a reaction (the number of transformation per unit time)?

Law of mass action:

the rate of a reaction is proportional to the product of the concentration, raised to the power of its stoichiometric coefficient, of the involved reactant

\[
\alpha A + \beta B \rightarrow \text{product}
\]

\[
R(c_A, c_B, x, t) = k c_A^\alpha(x, t) c_B^\beta(x, t), \quad r_A(x, t) = -\alpha R, \quad r_B(x, t) = -\beta R
\]

The strong hypothesis: dilution and well **mixed condition** for chemicals.
FAST REACTIONS: Invading reactant concentration

$x/\lambda$ - invading concentration field $c/c_0$ at time $t/\tau_a = 0.00$

$y/\lambda$ - $P_e = 0.07$

(juanesgroup.mit.edu/pdeanna)  pietrodeanna@gmail.com

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FAST REACTIONS: product concentration

\( \frac{x}{\lambda} - \text{product concentration field} \frac{c}{c_0} \text{ at time} \ t/\tau_a = 0.00 \)

\( \frac{y}{\lambda} - P_{e} = 0.07 \)

(juanesgroup.mit.edu/pdeanna)  pietrodeanna@gmail.com

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The filamentary structure of mixing and reaction kinetics

At the Darcy scale, assuming pore scale compete mixing

\[ \phi \frac{\partial \bar{c}_i(x, t)}{\partial t} + q \frac{\partial \bar{c}_i(x, t)}{\partial x} - \phi D^* \frac{\partial^2 \bar{c}_i(x, t)}{\partial x^2} = r_i^*(x, t) \]

\[ M_C(t) = \int_V c_C(\mathbf{x}, t) d\mathbf{x} \]

\[ M_C(t) = c_0 l_0 \phi \sqrt{\frac{4D^*t}{\pi}} \]

C. Gramiling et al. Env. Sci. & Tech. 2002


(juanesgroup.mit.edu/pdeanna)  pietrodeanna@gmail.com
The filamentary structure of mixing and reaction kinetics
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Pore scale: fast reactions \( \rightarrow \) kinetics is dominated by diffusive flux

\[ \frac{dM_C(t)}{dt} = D \int_{\Xi(t)} |\nabla c_A(\mathbf{x}, t)| d\Sigma = \frac{D\Sigma(t)c_0}{s}, \quad |\nabla c_A(\mathbf{x}, t)| \sim \frac{c_0}{s} \]

imposing volume conservation...

\[ \frac{1}{s(t)} \frac{ds(t)}{dt} = \frac{D}{s(t)^2} - \frac{1}{\Sigma(t)} \frac{d\Sigma(t)}{dt} \]


(juanesgroup.mit.edu/pdeanna) pietrodeanna@gmail.com
The filamentary structure of mixing and reaction kinetics

\[ \frac{1}{s(t)} \frac{ds(t)}{dt} = \frac{D}{s(t)^2} - \frac{1}{\Sigma(t)} \frac{d\Sigma(t)}{dt}, \quad \Sigma = \Sigma_0 (1 + \gamma t) \]

\[ s(t) = s_0 \sqrt{\frac{3\beta - 2 + 2(1 + \gamma t)^3}{3\beta (1 + \gamma t)^2}}, \quad \beta = \frac{\gamma s_0}{D} \]

The filamentary structure of mixing and reaction kinetics

\[ M_C \sim (t/\tau_a)^{1/2} \]

Fickian scaling - (t/\tau_a)^{1/2}

Pe = 0.07
Pe = 0.71
Pe = 2.4
Pe = 7.1


pietrodeanna@gmail.com
The filamentary structure of mixing and reaction kinetics

When lamellae are independent \( \Sigma = \Sigma_0(1 + \gamma t) \)...

...true until diffusion merges adjacent lamellae at \( t = \tau_m \).


(juanesgroup.mit.edu/pdeanna)  pietrodeanna@gmail.com  Peyresq - August 2014  26 / 45
The filamentary structure of mixing and reaction kinetics

When lamellae begin to coalescence the interface length is estimated by \( \Sigma = l_b n_b \)

The average bundle length is expected to be

\[
l_b \sim \sigma_a^2(t) \quad \text{and} \quad n_b \sim \frac{1}{s}, \quad \rightarrow \quad \frac{dM_C(t)}{dt} = \frac{D\Sigma c_0}{s} \propto \frac{Dc_0 \sigma_a(t)}{s(t)^2}
\]

For \( t > \tau_m \), \( s \sim \sqrt{t} \), thus \( \frac{dM_C(t)}{dt} \propto c_0 \sigma_a(t) \). Since \( \sigma_a(t) \sim t^{0.65} \), therefore \( M_C \sim \sigma_a(t) \).


(juanesgroup.mit.edu/pdeanna)  pietrodeanna@gmail.com
The filamentary structure of mixing and reaction kinetics


(juanesgroup.mit.edu/pdeanna)  pietrodeanna@gmail.com

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Experimental techniques:

porous media and visualization
Analogous porous media: Hele-Shaw cell
Microfluidics for porous media


(juanesgroup.mit.edu/pdeanna)  pietrodeanna@gmail.com

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Microfluidics for porous media


Number of publications in different categories from 2000 to 2012.

- Microfluidics in engineering journals
- Microfluidics in multidisciplinary journals
- Microfluidics in biology and medicine journals

Microfluidics for porous media - the flow

\[ Re = \frac{u\lambda}{\nu} \sim 0.005 \]

at \( Re \ll 1 \) the flow \( \nu \nabla^2 u = \nabla p \)
Microfluidics for porous media

\[ Re = \frac{u\lambda}{\nu} \sim 0.005 \]

at \( Re \ll 1 \) the flow \( \nu \nabla^2 u = \nabla p \)

\[ \lambda \gg a, \partial_x^2 \sim \partial_y^2 \ll \partial_z^2 \]

\[ \nabla^2 u \propto u_{xy} \text{ (Darcy)} \]

\[ \lambda \sim a = 100 \mu m, \text{ 3d Stokes flow} \]
Experimental techniques:

flow through porous media
Particle Image Velocimetry

It is a technique allowing us to quantify the local velocity field, by measuring the correlation between particle (or cloud of particles) positions.
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Particle Image Velocimetry

Let’s analyze this image with volume statistics...

Hp: each particle produces a nearly Gaussian image on a picture and they DON’T superpose.

\[ I(\mathbf{x}, t) = \tau(\mathbf{x}) * \sum_{i=1}^{N} I_0(\mathbf{x}_i) \delta(\mathbf{x} - \mathbf{x}_i) = \sum_{i=1}^{N} I_0(\mathbf{x}_i) \tau(\mathbf{x} - \mathbf{x}_i) \]
Particle Image Velocimetry

Particles image intensity

\[ I(x, t) = \sum_{i=1}^{N} I_0(x_i) \tau(x - x_i) \]

computing the 1–image single exposure auto-covariance

\[ R_1(s, t) = \langle I(x, t)I(x + s, t) \rangle = \]
Particle Image Velocimetry

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\[ R_1(s, t) = \langle I(x, t)I(x + s, t) \rangle = \]

\[ = \sum_{k=1}^{N} \left[ \sum_{i=1}^{N} I_0(x_i) \tau(x_k - x_i) \sum_{j=1}^{N} I_0(x_j) \tau(x_k - x_j + s) \right] = \]
Particle Image Velocimetry

Particles image intensity

\[ I(\boldsymbol{x}, t) = \sum_{i=1}^{N} I_0(\boldsymbol{x}_i) \tau(\boldsymbol{x} - \boldsymbol{x}_i) \]

computing the 1–image single exposure auto-covariance

\[ R_1(s, t) = \langle I(\boldsymbol{x}, t)I(\boldsymbol{x} + s, t) \rangle = \]

\[ = \sum_{k=1}^{N} \left[ \sum_{i=1}^{N} I_0(\boldsymbol{x}_i) \tau(\boldsymbol{x}_k - \boldsymbol{x}_i) \sum_{j=1}^{N} I_0(\boldsymbol{x}_j) \tau(\boldsymbol{x}_k - \boldsymbol{x}_j + s) \right] = \]

\[ = \sum_{k=1}^{N} \left[ \sum_{j \neq i}^{N} I_0(\boldsymbol{x}_i)^2 \tau(\boldsymbol{x}_k - \boldsymbol{x}_i) \tau(\boldsymbol{x}_k - \boldsymbol{x}_j + s) + \right. \]

\[ + \sum_{i=j}^{N} I_0(\boldsymbol{x}_i)^2 \tau(\boldsymbol{x}_k - \boldsymbol{x}_i) \tau(\boldsymbol{x}_k - \boldsymbol{x}_j + s) \]

The first term is zero (or negligible) and the second shows a peak at \( s = 0 \).
Particle Image Velocimetry

Particles image intensity

\[ I(x, t) = \sum_{i=1}^{N} I_0(x_i) \tau(x - x_i) \]

computing the 1–image single exposure auto-covariance using the Convolution Theorem...

\[ R_1(s, t) = \langle I(x, t)I(x + s, t) \rangle = \mathcal{F}^{-1}\left[ \mathcal{F}[I(x, t)]^* \mathcal{F}[I(x + s, t)] \right] \]

\[ = \sum_{k=1}^{N} \left[ \sum_{i=1}^{N} I_0(x_i) \tau(x_k - x_i) \sum_{j=1}^{N} I_0(x_j) \tau(x_k - x_j + s) \right] \]

\[ = \sum_{k=1}^{N} \left[ \sum_{j \neq i}^{N} I_0(x_i)^2 \tau(x_k - x_i) \tau(x_k - x_j + s) + \sum_{i=j}^{N} I_0(x_i)^2 \tau(x_k - x_i) \tau(x_k - x_j + s) \right] \]

The first term is zero (or negligible) and the second shows a peak at \( s = 0 \).

(juanesgroup.mit.edu/pdeanna)  pietrodeanna@gmail.com  Peyresq - August 2014  33 / 45
Particle Image Velocimetry

the peak position $s = 0$ represent the mean displacement of particles within the image

$$R_1(s, t) = \mathcal{F}^{-1}\left[\mathcal{F}[I(x, t)]^* \mathcal{F}[I(x + s, t)]\right]$$
Particle Image Velocimetry

the peak position $s = 0$ represent the mean displacement of particles within the image

$$R_1(s, t) = \mathcal{F}^{-1} \left[ \mathcal{F}[I(x, t)]^* \mathcal{F}[I(x + s, t)] \right]$$
Particle Image Velocimetry

Let’s now consider two images taken at a time difference $\Delta t$ such that the flow displace the particles of $d$

$$R_2(s, t|d) = \langle I_1(x, t) I_2(x + s + d, t + \Delta t) \rangle = \ldots$$

$$= \mathcal{F}^{-1} \left[ \mathcal{F}[I_1(x, t)]^* \mathcal{F}[I_2(x + s + d, t)] \right]$$
Particle Image Velocimetry

for a given displacement $d$ the peak is at $s = -d$

$$R_2(s, t|d) = \mathcal{F}^{-1}
\left[ \mathcal{F}[I_1(x, t)] \ast \mathcal{F}[I_2(x + s + d, t)] \right]$$
Particle Image Velocimetry

for a given displacement $d$ the peak is at $s = -d$

\[ R_2(s, t|d) = \mathcal{F}^{-1}\left[ \mathcal{F}[I_1(x, t)]^* \mathcal{F}[I_2(x + s + d, t)] \right] \]
Particle Image Velocimetry
Particle Image Velocimetry
Particle Image Velocimetry
Particle Image Velocimetry: \( u = \frac{d}{\Delta t} \)
Particle Image Velocimetry: $u = \frac{d}{\Delta t}$
Particle Image Velocimetry

4X vs 40X magnification with camera resolution of 2560 \times 2160 \text{px}^2

depth of the field of view $\sim 100\mu m$ vs $\sim 5\mu m$
Particle Image Velocimetry

Longitudinal velocity - $u_x$ (mm/s)
Particle Image Velocimetry

Longitudinal velocity - $u_x$ (mm/s)

axis ticks in mm

0 1 2 3 4 5 6 7 8

Longitudinal velocity - $u_x$ (mm/s)

(juanesgroup.mit.edu/pdeanna)  pietrodeanna@gmail.com

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PIV in another geometry

Axes in [mm] - Longitudinal velocity $u_x$ [mm/s]
PIV in another geometry
Experimental techniques:

transport through porous media
Transport of a conservative tracer: FLUORESCENCE

We dissolve in water fluorescein sodium salt: when excited by a radiation of wavelength $\lambda_{ex} = 494\text{nm}$ will emit a radiation of wavelength $\lambda_{em} = 521\text{nm}$.

![Diagram](image)

We produce a sharp front of fluorescent tracer.

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We calibrate the setup: observed light intensity vs. the tracer (known) concentration.
Transport of a conservative tracer: FLUORESCENCE

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We calibrate the setup: observed light intensity vs. the tracer (known) concentration.

(juanesgroup.mit.edu/pdeanna)  pietrodeanna@gmail.com  Peyresq - August 2014
Transport of a reactive tracer: chemiluminescence

We use two reactants that, when mixed, will start a chemiluminescent reaction.

Field of light intensity produced by chemiluminescent reactions at the reactants front

P. de Anna et al. Env. Sci & Tech. 2014
That’s all.